FORM PTO-1390 (Modified) (REV 11-98) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER: **GL-12 (GL-01-8)**

U.S APPLICATION NO. (IF KNOWN, SEE 37 CFR): **09/830841**

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/EP99/08388	**05 November 1999**	**06 November 1998**

TITLE OF INVENTION

LIQUID COMPOSITION POLYMERIZABLE INTO ORGANIC GLASSES HAVING GOOD OPTICAL AND PHYSICO-MECHANICAL PROPERTIES

APPLICANT(S) FOR DO/EO/US

Fiorenzo Renzi, Andrea Bendandi, Roberto Forestieri and Nereo Nodari

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
   a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☒ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☐ A copy of the International Search Report (PCT/ISA/210).
8. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☒ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☐ have not been made and will not be made.
9. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
10. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
11. ☒ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).

Items 13 to 20 below concern document(s) or information included:

13. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☒ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☒ Certificate of Mailing by Express Mail
20. ☒ Other items or information:

Form PTO/IB/308

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
09/830841	PCT/EP99/08388	GL-12 (GL-01-8)

21. The following fees are submitted:.

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :

		CALCULATIONS PTO USE ONLY
☐	Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2) paid to USPTO and International Search Report not prepared by the EPO or JPO	**$1,000.00**
☒	International preliminary examination fee (37 CFR 1.482) not paid to USPTO but Internation Search Report prepared by the EPO or JPO	**$860.00**
☐	International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO	**$710.00**
☐	International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4)	**$690.00**
☐	International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4)	**$100.00**
	ENTER APPROPRIATE BASIC FEE AMOUNT =	**$860.00**
	Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).	**$0.00**

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	25 - 20 =	5	x $18.00	**$90.00**
Independent claims	3 - 3 =	0	x $80.00	**$0.00**
Multiple Dependent Claims **(check if applicable)**.			☐	**$0.00**
	TOTAL OF ABOVE CALCULATIONS =			**$950.00**
	Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) **(check if applicable)**. ☐			**$0.00**
	SUBTOTAL =			**$950.00**
	Processing fee of **$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (f)). +			**$0.00**
	TOTAL NATIONAL FEE =			**$950.00**
	Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) **(check if applicable).** ☐			**$0.00**
	TOTAL FEES ENCLOSED =			**$950.00**
				Amount to be: refunded $
				charged $

☐ A check in the amount of to cover the above fees is enclosed.

☒ Please charge my Deposit Account No. **50-0935** in the amount of **$950.00** to cover the above fees.
A duplicate copy of this sheet is enclosed.

☒ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. **50-0935** A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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REGISTRATION NUMBER

May 1, 2001
DATE

09/830841

CERTIFICATE OF MAILING BY "EXPRESS MAIL" (37 CFR 1.10)

Applicant(s): **Fiorenzo Renzi et al.**

0608 Rec'd PCT/PTO 01 MAY 2001

Docket No. **GL-12 (GL-01-8)**

Serial No.	Filing Date	Examiner	Group Art Unit
UNKNOWN	**HEREWITH**	**UNKNOWN**	**UNKNOWN**

Invention: **LIQUID COMPOSITION POLYMERIZABLE INTO ORGANIC GLASSES HAVING GOOD OPTICAL AND PHYSICO-MECHANICAL PROPERTIES**

I hereby certify that the following correspondence:

National Phase Pat. Appln., IPER (Form PCT/IPEA/416 & 409), Form PCT/IB308 & return postcard
+ Declaration (executed)

(Identify type of correspondence)

is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 in an envelope addressed to: The Assistant Commissioner for Patents, Washington, D.C. 20231 on

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(Date)

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(Typed or Printed Name of Person Mailing Correspondence)

Michael W. Ferrell
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IN THE UNITED STATE PATENT AND TRADEMARK OFFICE
(UNITED STATES RECEIVING OFFICE)

In re Application of:	:	
Fiorenzo Renzi et al.	:	
U.S. Serial No. UNKNOWN	:	Group Art Unit: UNKNOWN
Filed HEREWITH	:	
Docket No. GL-12 (GL-01-8)	:	Examiner: UNKNOWN
Title: LIQUID COMPOSITION POLYMERIZABLE INTO ORGANIC GLASSES HAVING GOOD OPTICAL AND PHYSICO-MECHANICAL PROPERTIES	:	

Box PCT
Assistant Commissioner for Patents
Washington, D.C. 20231

PRELIMINARY AMENDMENT

Sir:

Prior to calculation of the filing fee, please enter the following amendments:

IN THE CLAIMS

Please amend claims 4, 6, 9, 11, 13, 15, 16 21, 22 and 23 to read as follows:

4. The composition according to claim 1, wherein the polyols (c) are: pentaerythritol, trimethylolpropane, dipentaerythritol, ditrimethylolpropane, tris(hydroxyethyl) isocyanurate.

6. The composition according to claim 1, obtained starting from diallyl carbonate (A) and from the mixture (B+C) operating under transesterification conditions, at a temperature ranging from 80°C to 160°, in the presence of a catalyst of the alkaline type, and continuously eliminating the allyl alcohol which is formed as reaction by-product.

9. The composition according to claim 6, wherein the catalyst is used in a quantity equal to at least 1 ppm (parts per million by weight) with respect to the sum of the weights of components (B+C).

11. The composition according to claim 6, wherein the transesterification reaction is carried out at pressure values ranging form 60 mbar to 1030 mbar.

13. The composition according to claim 6, wherein the reaction times range from 0.5 hours to 20 hours.

15. The composition according to claim 1, wherein one or more conventional additives are present, such as oxidization, light and heat stabilizers, lubricants, dyes, pigments, UV-absorbers, IR-absorbers, and the like, in a total quantity however not exceeding 1 part by weight for every 100 parts by weight of the compositions themselves.

16. The composition according to claim 1, wherein one or more polymerization initiators are present, which are soluble in the composition itself and are capable of generating free radicals within a temperature range of 30°C to 120°C.

21. The composition according to claim 16, wherein the quantity of initiator used varies within a range of 1 to 6 parts by weight for every 100 parts by weight of said composition.

22. The composition according to claim 16, which is transformed into the relative organic glasses operating at a temperature ranging form 30°C to 120°C, with polymerization times which generally range from 1 hour to 100 hours.

23. Organic glasses obtained from the polymerization of the composition according to claim 1.

REMARKS

The foregoing amendment is made to eliminate multiple dependencies. For purposes of calculating the filing fee, there are twenty-five (25) claims, twenty-two (22) being dependent and a filing fee of $950.00 is warranted. The foregoing amendment is made on the basis that the amendments to the case indicate on the Examination Report have been previously entered. The changes to the claims are shown on the attached marked-up pages.

Respectfully submitted,

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4. The composition according to ~~any of the previous claims~~claim 1, wherein the polyols (c) are: pentaerythritol, trimethylolpropane, dipentaerythritol, ditrimethylolpropane, tris(hydroxyethyl) isocyanurate.

6. The composition according to ~~any of the previous claims~~claim 1, obtained starting from diallyl carbonate (A) and from the mixture (B+C) operating under transesterification conditions, at a temperature ranging from 80°C to 160°, in the presence of a catalyst of the alkaline type, and continuously eliminating the allyl alcohol which is formed as reaction by-product.

9. The composition according to ~~any of the claims from 6 to 8~~claim 6, wherein the catalyst is used in a quantity equal to at least 1 ppm (parts per million by weight) with respect to the sum of the weights of components (B+C).

11. The composition according to ~~any of the claims from 6 to 10~~claim 6, wherein the transesterification reaction is carried out at pressure values ranging form 60 mbar to 1030 mbar.

13. The composition according to ~~any of the claims from 6 to 12~~claim 6, wherein the reaction times range from 0.5 hours to 20 hours.

15. The composition according to ~~any of the previous claims~~claim 1, wherein one or more conventional additives are present, such as oxidization, light and heat stabilizers, lubricants, dyes, pigments,

UV-absorbers, IR-absorbers, and the like, in a total quantity however not exceeding 1 part by weight for every 100 parts by weight of the compositions themselves.

16. The composition according to ~~any of the previous claims~~claim 1, wherein one or more polymerization initiators are present, which are soluble in the composition itself and are capable of generating free radicals within a temperature range of 30°C to 120°C.

21. The composition according to ~~any of the claims from 16 to 20~~claim 16, wherein the quantity of initiator used varies within a range of 1 to 6 parts by weight for every 100 parts by weight of said composition.

22. The composition according to ~~any of the claims from 16 to 21~~claim 16, which ~~are~~ 's transformed into the relative organic glasses operating at a temperature ranging form 30°C to 120°C, with polymerization times which generally range from 1 hour to 100 hours.

23. Organic glasses obtained from the polymerization of the composition according to ~~any of the previous claims~~claim 1.

LIQUID COMPOSITION POLYMERIZABLE INTO ORGANIC GLASSES HAVING GOOD OPTICAL AND PHYSICO-MECHANICAL PROPERTIES.

The present invention relates to a liquid composition polymerizable into organic glasses.

More specifically, the present invention relates to a liquid composition which can be polymerized, by means of radical polymerization with low shrinkage, into organic glasses having good optical and physico-mechanical properties, comprising the product obtained from the transesterification of a diallyl carbonate (A) with a mixture of one or more diols (B) with a polyol (C).

A further object of the present invention relates to the organic glasses obtained from the polymerization of said composition.

Finally, the present invention also relates to the end-articles obtained starting from said composition, such as for example, ophthalmic lenses and solar filters, protective shields, sight windows, solar and photovoltaic collectors and panels, substrates for optical disks, panels

for display and video terminals.

In the field of high transparency organic glasses, the product obtained from the polymerization of diethylene glycol bis(allyl carbonate) is of great commercial interest in the production of ophthalmic and safety plates and lenses, owing to its special mechanical and age-resistance characteristics, as described, for example, by F. Strain, in: "Encyclopedia of Chemical Processing and Design", First Edition, Dekker Inc., New York, Vol. 11, page 452 onwards; and in "Encyclopedia of Polymer Science and Technology" (1964), Vol. 1, page 799 onwards, Interscience Publishers, New York.

The use of diethylene glycol bis(allyl carbonate), however, has various disadvantages which limit and, at times, prevent its use in different fields of application.

For example, the shrinkage which accompanies the polymerization reaction of bis (allyl carbonate) in the presence of peroxide initiators, makes the preparation of high power lenses difficult. The abrasion resistance of the organic glasses thus obtained, although much higher than that of other known organic glasses, still cannot be considered as being optimum: this is demonstrated by the fact that it is customary to resort to the surface application of scratch-proof coatings on this organic glass.

Not even the impact strength of the above organic

glasses, although its value is sufficient to pass the tests imposed by the regulations in force in the optical field, can be considered as being optimum.

Numerous compositions polymerizable into organic glasses have been described in an effort to overcome these drawbacks and to improve some of the most important characteristics of organic glasses.

For example, the patent U.S. 4,812,545, discloses liquid compositions polymerizable into organic glasses comprising tris(hydroxyethyl)isocyanurate tris(allyl carbonate) and diethylene glycol bis(allyl carbonate) monomeric and oligomeric, having an improved shrinkage during polymerization and a better impact strength.

The patent U.S. 4,713,433 on the other hand, describes liquid compositions polymerizable into organic glasses comprising oligomeric bis(allyl carbonate) and a comonomer having at least four terminal allyl groups capable of producing organic glasses with an improved abrasion resistance.

Finally, the patent U.S. 4,970,293 discloses liquid compositions polymerizable into organic glasses comprising the reaction product of a diallyl carbonate with mixtures of a diol and a polyol containing from three to six hydroxyl groups per molecule. However, although these compositions on the one hand effectively represent an improve-

ment in some specific characteristics of the organic glasses obtained from their polymerization, on the other hand, they have other characteristics which are worse than those of the organic glasses obtained from the polymerization of diethylene glycol bis(allyl carbonate) alone.

As already mentioned above, the organic glass obtained from the polymerization of diethylene glycol bis(allyl carbonate) alone has various disadvantages: for example, it often has yellow index values higher than those normally acceptable, and/or unacceptable refraction index values, and/or impact strenght values, and/or dyeability values. As a result, diethylene glycol bis(allyl carbonate) cannot be used alone but, as specified above, must be mixed with other comonomers which, however, have other drawbacks.

In this respect, it should be remembered that the organic glass obtained as described in the patent U.S. 4,970,293 mentioned above, is particularly useful in protective shields (for example, for welders), in sight windows (for example, in blast furnaces), in windows in the transport and civil industry, in lenses for vehicle lights, in solar and photovoltaic collectors and panels, in substrates for optical disks and in panels for display, but it cannot be used for optical lenses as it has a high yellow index, a low impact strenght, a poor dyeability.

The Applicant has now found a liquid composition which

can be easily polymerized by means of radical polymerization with low shrinkage, into organic glasses having good optical and physico-mechanical properties, capable of overcoming the drawbacks of the known art described above.

The present invention therefore relates to a liquid composition which can be polymerized by means of radical polymerization with low shrinkage, into organic glasses, comprising the product obtained from the transesterification of a diallyl carbonate (A) with a mixture of one or more linear or branched aliphatic diols (B), containing from three to ten carbon atoms in the molecule with a linear or branched aliphatic polyol (C), containing from four to twenty carbon atoms and from three to six hydroxyl groups in the molecule, wherein

~~In the liquid composition of the present invention, the molar ratio A/(B+C) ranges from 2/1 to 5/1 and the quantity of (C) in the mixture (B+C) is equal to or less than 25% by weight with respect to the total weight of the mixture (B+C).~~

~~In the liquid composition of the present invention,~~ the molar ratio (A)/(B+C) ~~preferably~~ ranges from 2.5/1 to 4/1, and the quantity of (C) in the mixture (B+C) ranges from 5% by weight to 20% by weight with respect to the total weight of the mixture (B+C).

Diols (B) which can be used for the purposes of the

present invention, as already mentioned above, are linear or branched aliphatic diols, containing from three to ten carbon atoms in the molecule.

Specific examples of diols (B) which can be used for the purposes of the present invention are: diethylene glycol, triethylene glycol, tetraethylene glycol, 1,4-butanediol, 1,6-hexanediol, 1,3-propanediol, neopentylglycol, dipropyleneglycol, 2,2,4-trimethyl-1,3-pentanediol, etc.

Preferred diols for the purposes of the present invention are diethylene glycol and neopentylglycol.

Polyols (C) which can be used for the purposes of the present invention, as already mentioned above, are linear or branched aliphatic polyols, containing from four to twenty carbon atoms and from three to six hydroxyl groups in the molecule.

Specific examples of polyols (C) which can be used for the purposes of the present invention are: pentaerythritol, trimethylolpropane, dipentaerythritol, ditrimethylolpropane, tris(hydroxyethyl)isocyanurate, etc.

Preferred polyols for the purposes of the present invention are pentaerythritol and trimethylolpropane.

The polymerizable liquid composition of the present invention is obtained starting from diallyl carbonate (A) and the mixture (B+C) operating under transesterification conditions. More specifically, the reagents are put in con-

tact with each other, in the ratios indicated above, and reacted at a temperature ranging from 80°C to 160°C, preferably from 90°C to 130°C, in the presence of a catalyst of the alkaline type, continuously eliminating the allyl alcohol formed as reaction by-product.

Catalysts of the alkaline type which can be used for the purposes of the present invention are: hydroxides, carbonates and alcoholates of alkaline metals, organic bases, basic ion-exchange resins.

Specific examples of catalysts of the alkaline type used for the purposes of the present invention are: sodium hydroxide, sodium carbonate, sodium methylate.

The catalyst is conveniently used in a quantity equal to at least 1 ppm (parts per million by weight) with respect to the sum of the weights of components (B+C) and, preferably, in a quantity ranging from 0.01% to 0.3% by weight.

The above transesterification reaction is conveniently carried out at such a pressure as to bring the system to boiling point at the preselected operating temperature, in order to favour the elimination of the allyl alcohol from the reaction mixture: for example, pressure values ranging from 60 mbar to 1030 mbar, preferably from 60 mbar to 500 mbar, are suitable for the purpose.

Operating under the conditions described above, the

reaction times generally range from 0.5 hours to 20 hours, preferably from 0.5 hours to 3 hours.

After cooling, the above reaction mixture is washed with water to remove the small quantities of residual catalyst and, after separation and removal of the aqueous phase, the non-reacted diallyl carbonate is eliminated by distillation, heating to a temperature in the order of 130°C, under decreasing pressure with end-values ranging from 0.1 mbar to 20 mbar, preferably from 0.5 mbar to 2 mbar, obtaining the desired composition, as residue.

The composition thus obtained is finally subjected to filtration after optional treatment with activated carbon.

The composition of the present invention is liquid at room temperature and has viscosity values ranging from 15 cts to 300 cts and density values ranging from 1.1 g/ml to 1.3 g/ml.

The polymerizable liquid composition of the present invention is a complex mixture which contains allyl carbonates of component (B) and component (C), in monomeric and oligomeric form, as well as mixed oligomeric allyl carbonates of these compounds (B) and (C), the relative quantities of these constituents of the present composition mainly depending on the pre-established ratios of reagents (A), (B) and (C).

The above composition can be transformed into organic

glasses, by means of radical polymerization, using the usual "casting" technique.

For this purpose, one or more polymerization initiators are added to the composition, which are soluble in the composition itself and capable of generating free radicals within a temperature range of 30°C to 120°C.

A group of polymerization initiators which can be used for the purposes of the present invention is the group of peroxides.

Preferred examples of peroxides which can be used for the present invention are: dicyclohexylperoxydicarbonate, diisopropylperoxydicarbonate, dibenzoylperoxide, di-s-butyl-peroxydicarbonate, s-butylcyclohexylperoxydicarbonate, etc.

Other peroxides which can be used for the purposes of the present invention are perketals.

Preferred examples of perketals which can be used in the present invention are: 1,1-di-(t-butylperoxy)-cyclohexane, 1,1-di-(t-butylperoxy)-3,3,5-trimethyl-cyclohexane, 1,1-di-(t-amylperoxy)-cyclohexane, 1,1-di-(t-butylperoxy)-2-methylcyclohexane, 1,1-di-(t-amylperoxy)-2-methylcyclohexane, etc.

The quantity of initiator used may generally vary within a range of 1 to 6 parts by weight for every 100 parts by weight of the composition of the present inven-

tion.

The composition of the present invention may optionally contain one or more conventional additives such as, for example, oxidization, light and heat stabilizers, lubricants, dyes, pigments, UV-absorbers, IR-absorbers, and the like, in a total quantity however not exceeding 1 part by weight for every 100 parts by weight of the compositions themselves.

Examples of additives which can be used for the purposes of the present invention are: sterically hindered phenols, sterically hindered amines, benzophenones, benzotriazoles, organic phosphites and phosphonites, etc.

The composition of the present invention containing the polymerization initiator and, optionally, one or more additives selected from those mentioned above, is transformed into the relative organic glasses, operating at temperatures ranging from 30°C to 120°C, with polymerization times which can generally vary from 1 hour to 100 hours.

During the polymerization there is a limited shrinkage and the organic glasses thus obtained have good optical and physico-mechanical properties.

These organic glasses are particularly useful in the production of ophthalmic lenses and solar filters, protective shields, sight windows, solar and photovoltaic collectors and panels, substrates for optical disks, panels for

display and video terminals: these end-articles are therefore a further object of the present invention.

Some illustrative examples are provided for a better understanding of the present invention and for its embodiment, but should not be considered as limiting the scope of the invention in any way.

In the following examples polymerizable liquid compositions are prepared by reacting, under transesterification conditions, the diallyl carbonate (A) and a mixture of compounds (B) and (C).

The diol (B) used in the examples is diethylene glycol (DEG).

The polyol (C) used in the examples is pentaerythritol (PE).

Dicyclohexylperoxydicarbonate (CHPC) is added to the liquid compositions thus obtained, as polymerization initiator, in a quantity equal to 5% by weight with respect to the weight of the composition.

The compositions containing the polymerization initiator are transformed, by means of polymerization, into flat sheets or neutral lenses, using the "casting" technique. Operating according to this technique, the liquid compositions, containing the polymerization initiator, are poured into the cavity of a mould consisting of two glass elements and having a spacer gasket of plasticized polyvinylchlo-

ride, of ethylene-vinylacetate (EVA) copolymer, of low density polyethylene (LDPE), or of any other suitable material, compatible with the operating conditions.

The liquid compositions are then subjected to polymerization by means of thermal treatment in a forced circulation oven, with a gradual temperature rise from 35°C to 80°C in twenty hours.

At the end of the above treatment, the moulds are opened and the polymerized products are recovered and maintained at 110°C for an hour to complete the polymerization reaction and give the end-article dimensional stability.

The following characteristics are determined on the sheets thus obtained:

(a) Optical characteristics

- Refractive index (n_D^{20}): measured with an Abbe refractometer (ASTM D-542).
- Yellow index (YI) (ASTM D-1925) defined as

$$YI = \frac{100}{Y} \quad (1.277X - 1.06Z)$$

determined with a Macbeth 1500 Plus spectrophotometer.

(b) Physical and mechanical characteristics

- Density: determined with hydrostatic balance at a temperature of 20°C (ASTM D-792).
- Shrinkage during polymerization calculated with the following formula:

$$\% \text{ shrinkage} = \frac{(\text{polymer density} - \text{monomer density})}{(\text{polymer density})} \times 100$$

- Rockwell Hardness (M) measured with a Rockwell durometer (ASTM D-785).
- Izod impact strenght without notch (ASTM D-256 modified)

(c) Thermal characteristics

- Deflection temperature under load 1.82 Mpa (HDT) (ASTM D-648).

(d) Abrasion resistance

To evaluate the abrasion resistance the Sutherland rub tester is used. The test consists in carrying out 50 passages with a 2/0 type steel wool bearing loaded with a weight of 2 kg on a neutral sample lens.

The abrasion degree produced is evaluated by measuring the Haze % increase (% of diffused light transmitted with respect to the total light transmitted) following scratches produced on the surface of the lens.

The Haze values are determined before and after the abrasion test using a Hazegard XL-211 device of Gardner, in accordance with the regulation ASTM D-1003.

A higher Haze value obviously indicates a greater degree of diffused light transmitted and consequently a lower abrasion resistance (more scratched lens).

(e) Dyeability

The capacity to adsorb a dye on the surface of the material is determined by the immersion of a neutral lens in an aqueous bath in which the dye BPI Guy is dispersed.

For this purpose the lens is immersed in this colouring bath for 30 minutes at a temperature of 80°C and, after rinsing with demineralized water, the light transmittance of the lens is determined by measuring the Y chromatic coordinate as described by CIE (1931) Standard Observer.

It can be clearly seen from the following examples that the compositions of the present invention, as well as having a reduced shrinkage during polymerization, allow the production of organic glasses having improved characteristics with respect to the organic glasses of the known art:

- refractive index equal to that of the organic glasses obtained from the polymerization of diethylene glycol bis(allyl carbonate) alone (this allows the use of the same glass moulds with a consequent reduction in the investment costs);
- reduced yellow index;
- high impact strenght;
- high abrasion resistance;
- good dyeability.

EXAMPLE 1

The following products are charged into a three-

necked, jacketed flask, equipped with a thermometer and magnetic stirrer and overhead with a distillation column with 10 perforated trays having a diameter of 30 mm:

- pentaerythritol (PE): 34.5 g (0.25 moles);
- diethylene glycol (DEG): 223 g (2.10 moles);
- diallyl carbonate (DAC): 1000 g (7.04 moles);
- solution at 20% by weight of sodium methylate in methanol (1.20 ml).

The reaction is carried out for 3 hours at a temperature of 83°C-120°C and at a pressure decreasing from 190 mbar to 130 mbar, and distilling the allyl alcohol as it is formed (total 346 ml; purity > 99.0%).

After cooling, the reaction mixture is washed with two portions, each of 500 ml, of distilled water.

The excess of diallyl carbonate is distilled at a pressure of about 1 mbar, operating at a temperature increasing up to 130°C: the product obtained is filtered with a 0.45 μm membrane.

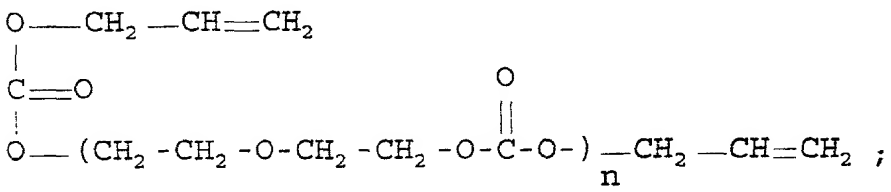
525 g of a liquid product are thus obtained, having the following characteristics:

- viscosity (25°C): 75 cst;
- density (20°C): 1.192 g/ml;
- refractive index (n_D^{20}): 1.461;
- APHA colour: 5

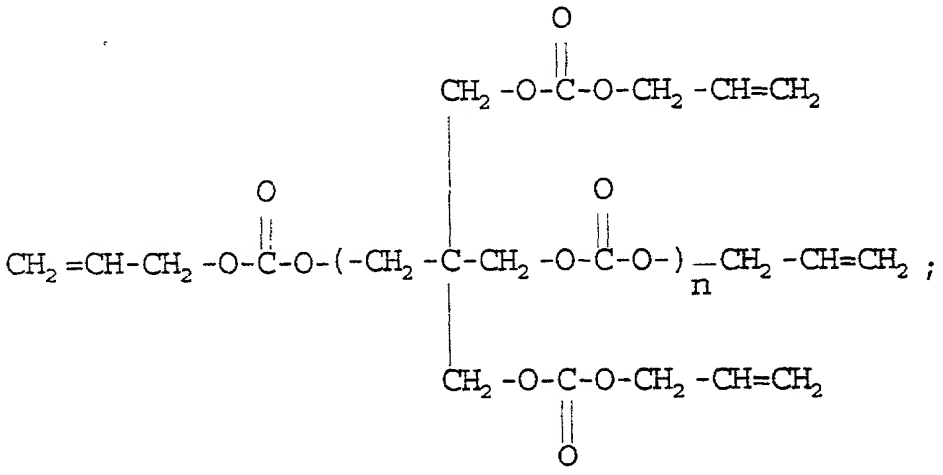
The above product is a mixture of diethylene glycol

bis(allyl carbonate) monomer and oligomers, of pentaerythritol tetrakis(allyl carbonate) monomer and oligomers, and mixed allyl carbonates, having the following composition determined by liquid column chromatography (HPLC; ELDS detector):

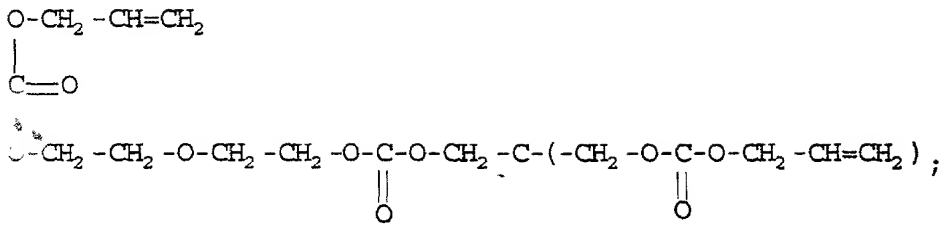
- 80% by weight of diethylene glycol bis(allyl carbonate) monomer (n = 1) and oligomers (n > 1), having the formula:



- 3% by weight of pentaerythritol tetrakis(allyl carbonate), monomer (n = 1) and oligomers (n > 1), having the formula:



- 12% by weight of mixed allyl carbonate having the formula:



the remaining percentage essentially consisting of higher oligomers of the previous species.

The above composition, after the addition of dicyclohexylperoxydicarbonate (CHPC; 5% by weight in the composition), is subjected to polymerization operating as described above, and the characteristics indicated in Table 1 are determined on the hardened composition.

Table 1 indicates, for comparative purposes, the properties of the composition obtained from the polymerization of diethylene glycol bis(allyl carbonate) alone and the composition obtained from the polymerization of the liquid composition described in Example 3 of the patent U.S. 4,970,293: the polymerization conditions are obviously the same as those used for the composition object of the present invention above described.

The yellow index (YI) is determined on a sample having a thickness of 5 mm to which 2-hydroxy-4-methoxy-benzophenone has been added (0.1%).

TABLE 1

	Composition Example 1	Diethylene glycol bis(allyl carbonate)	Composition Example 3 of U.S. 4,970,293
Density (20°C)	1.328	1.311	1.333
Shrinkage (%)	10.2	12.1	10.5
Refractive index (n_D^{20})	1.500	1.500	1.502
Yellow index (YI)	1.6	1.7	2.9
Rockwell Hardness (M)	100	98	116
Izod impact strenght without notch (KJ/m^2)	30	25	14
HDT (°C)	61	62	143
Sutherland abrasion resistance (Haze%)	0.4	1.0	0.2
Dyeability (Y)	42.9	38.3	76.2

EXAMPLE 2

Operating as described in Example 1, the following compositions 2, 3 and 4 are prepared, by reacting diallyl carbonate (DAC) with mixtures of pentaerythritol (PE) and diethylene glycol (DEG), in various molar ratios (DAC/(PE + DEG), as indicated in Table 2.

Table 2 also indicates the viscosity (cst; 25°C), density (g/ml; 20°C), and refraction index (n_D^{20}) characteristics of the polymerizable liquid compositions obtained.

TABLE 2

Composition Nr.		2	3	4
Mixture	PE (weight %)	11.6	12	11
	DEG (weight %)	88.4	88	89
DAC/(PE + DEG)		2.7/1	3.2/1	3.4/1
Viscosity (cst; 25°C)		92.3	61	56
Density (g/ml; 20°C)		1.194	1.190	1.187
n_D^{20}		1.461	1.460	1.459

The above compositions, after the addition of dicyclohexylperoxydicarbonate (CHPC; 5% by weight in the composition), are subjected to polymerization operating as described above and the characteristics indicated in Table 3 are determined on the hardened compositions.

Also in this case, the yellow index (YI) is determined on a sample having a thickness of 5 mm and to which 2-hydroxy-4-methoxybenzophenone has been added (0.1%).

TABLE 3

	Composition Nr. 2	Composition Nr. 3	Composition Nr. 4
Density (20°C)	1.326	1.325	1.350
Shrinkage (%)	9.9	10.2	10.4
Refractive index (n_D^{20})	1.499	1.500	1.500
Yellow index (YI)	1.6	1.6	1.7
Rockwell Hardness (M)	95	95	100
Izod impact strenght without notch (KJ/m^2)	39	34	42
HDT (°C)	-	60	61
Sutherland abrasion resistance (Haze%)	0.4	0.35	0.45
Dyeability (Y)	39.8	41.8	40.2

CLAIMS

1. A liquid composition which can be polymerized, by means of radical polymerization with low shrinkage, into organic glasses, comprising the product obtained from the transesterification of a diallylcarbonate (A) with a mixture of one or more linear or branched aliphatic diols (B), containing from three to ten carbon atoms in the molecule with a linear or branched aliphatic polyol (C), containing from four to twenty carbon atoms and from three to six hydroxyl groups in the molecule,

~~2. The composition according to claim 1, wherein the molar ratio A/(B+C) ranges from 2/1 to 5/1 and the quantity of (C) in the mixture (B+C) is equal to or less than 25% by weight with respect to the total weight of said mixture (B+C).~~

~~3. The composition according to claim 2,~~ wherein the molar ratio (A)/(B+C) ranges from 2.5/1 to 4/1 and the quantity of (C) in the mixture (B+C) ranges from 5% by weight to 20% by weight with respect to the total weight of said mixture (B+C).

2 4. The composition according to claim 1, ~~2 or 3,~~ wherein the diols (B) are: diethylene glycol, triethylene glycol, tetraethylene glycol, 1,4-butanediol, 1,6-hexanediol, 1,3-propanediol, neopentylglycol, dipropyle-

neglycol, 2,2,4-trimethyl-1,3-pentanediol.

3. The composition according to claim 2, wherein the diols are diethylene glycol and neopentylglycol.

4. The composition according to any of the previous claims, wherein the polyols (C) are: pentaerythritol, trimethylolpropane, dipentaerythritol, ditrimethylolpropane, tris(hydroxyethyl)isocyanurate.

5. The composition according to claim 4, wherein the polyols are pentaerythritol and trimethylolpropane.

6. The composition according to any of the previous claims, obtained starting from diallyl carbonate (A) and from the mixture (B+C) operating under transesterification conditions, at a temperature ranging from 80°C to 160°C, in the presence of a catalyst of the alkaline type, and continuously eliminating the allyl alcohol which is formed as reaction by-product.

7. The composition according to claim 6, wherein the transesterification is carried out at a temperature ranging from 90°C to 130°C, and the catalyst of the alkaline type is selected from: hydroxides, carbonates and alcoholates of alkaline metals, organic bases, basic ion-exchange resins.

8. The composition according to claim 7, wherein the catalyst of the alkaline type is selected from: sodium hydroxide, sodium carbonate, sodium methylate.

9 ~~11~~. The composition according to any of the claims from 6 ~~8~~ to 8 ~~10~~, wherein the catalyst is used in a quantity equal to at least 1 ppm (parts per million by weight) with respect to the sum of the weights of components (B+C).

10 ~~12~~. The composition according to claim 9 ~~11~~, wherein the catalyst is used in a quantity ranging from 0.01% to 0.3% by weight with respect to the sum of the weights of components (B+C).

11 ~~13~~. The composition according to any of the claims from 6 ~~8~~ to 10 ~~12~~, wherein the transesterification reaction is carried out at pressure values ranging from 60 mbar to 1030 mbar.

12 ~~14~~. The composition according to claim 11 ~~13~~, wherein the transesterification reaction is carried out at pressure values ranging from 60 mbar to 500 mbar.

13 ~~15~~. The composition according to any of the claims from 6 ~~8~~ to 12 ~~14~~, wherein the reaction times range from 0.5 hours to 20 hours.

14 ~~16~~. The composition according to claim 13 ~~15~~, wherein the reaction times range from 0.5 hours to 3 hours.

15 ~~17~~. The composition according to any of the previous claims, wherein one or more conventional additives are present, such as oxidization, light and heat stabilizers, lubricants, dyes, pigments, UV-absorbers, IR-

absorbers, and the like, in a total quantity however not exceeding 1 part by weight for every 100 parts by weight of the compositions themselves.

16
~~18~~. The composition according to any of the previous claims, wherein one or more polymerization initiators are present, which are soluble in the composition itself and are capable of generating free radicals within a temperature range of 30°C to 120°C.

17 16
~~19~~. The composition according to claim ~~18~~, wherein the polymerization initiators belong to the group of peroxides.

18 17
~~20~~. The composition according to claim ~~19~~, wherein the peroxides are: dicyclohexylperoxydicarbonate, diisopropylperoxydicarbonate, dibenzoylperoxide, di-s-butyl-peroxydicarbonate, s-butyl-cyclohexylperoxydicarbonate.

19 16
~~21~~. The composition according to claim ~~18~~, wherein the polymerization initiators are perketals.

20 19
~~22~~. The composition according to claim ~~21~~, wherein the perketals are: 1,1-di-(t-butylperoxy)-cyclohexane, 1,1-di-(t-butylperoxy)-3,3,5-trimethyl-cyclohexane, 1,1-di-(t-amyl-peroxy)-cyclohexane, 1,1-di-(t-butyl-peroxy)-2-methyl-cyclohexane, 1,1-di-(t-amylperoxy)-2-methylcyclohexane.

21 16
~~23~~. The composition according to any of the claims from ~~18~~

to 20 ~~22~~, wherein the quantity of initiator used varies within a range of 1 to 6 parts by weight for every 100 parts by weight of said composition.

22 16
~~24~~. The composition according to any of the claims from ~~18~~ to 21 ~~23~~, which are transformed into the relative organic glasses operating at a temperature ranging from 30°C to 120°C, with polymerization times which generally range from 1 hour to 100 hours.

23
~~25~~. Organic glasses obtained from the polymerization of the compositions according to any of the previous claims.

24
~~26~~. Ophthalmic lenses and solar filters, protective shields, sight windows, solar and photovoltaic collectors and panels, substrates for optical disks, panels for display, video terminals obtained from the processing of the organic glasses according to claim 23 ~~25~~.

25. **Use of a** liquid composition which can be polymerized, by means of radical polymerization with low shrinkage, into organic glasses, comprising the product obtained from the transesterification of a diallylcarbonate (A) with a mixture of one or more linear or branched aliphatic diols (B), containing from three to ten carbon atoms in the molecule with a linear or branched aliphatic polyol (C), containing from four to twenty carbon atoms and from three to six hydroxyl groups in the molecule, wherein the molar ratio (A)/(B+C) ranges from 2.5/1 to 4/1 and the quantity of (C) in the mixture (B+C) ranges from 5% by weight to 20% by weight with respect to the total weight of said mixture (B+C),
for manufacturing optical lenses.

Docket No.

Declaration and Power of Attorney For Patent Application

English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

LIQUID COMPOSITION POLYMERIZABLE INTO ORGANIC GLASSES HAVING GOOD OPTICAL AND PHYSICO-MECHANICAL PROPERTIES

☐ the specification of which

(check one)

☐ is attached hereto.

☒ was filed on 5 NOVEMBER 1999 as United States Application No. or PCT International Application Number PCT/EP99/08388

and was amended on _____ (if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) or Section 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)			Priority Not Claimed
MI98A002411	ITALY	6 NOVEMBER 1998	☒
(Number)	(Country)	(Day/Month/Year Filed)	
			☐
(Number)	(Country)	(Day/Month/Year Filed)	
			☐
(Number)	(Country)	(Day/Month/Year Filed)	

I hereby claim the benefit under 35 U.S.C. Section 119(e) of any United States provisional application(s) listed below:

(Application Serial No.)	(Filing Date)

I hereby claim the benefit under 35 U. S. C. Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. Section 112, I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, C. F. R., Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. *(list name and registration number)*

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Sixth inventor's signature — Date

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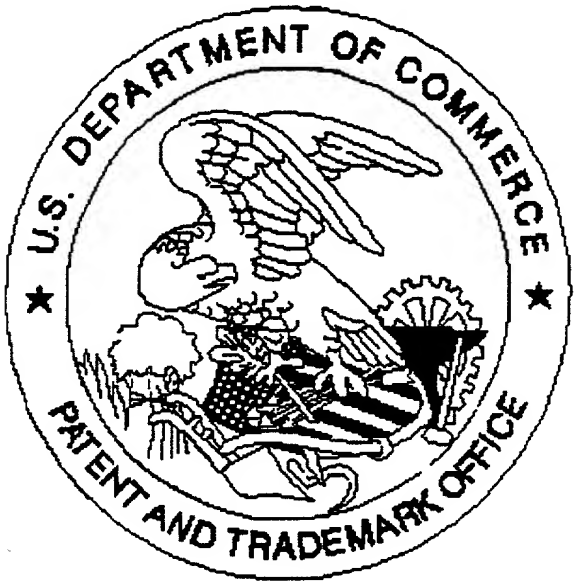

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Application papers not suitable for publication

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